MINISTRY OF AGRICULTURE, FISHERIES AND FOOD

Survey of styrene levels in food contact materials and in foods

The Eleventh Report of the
Steering Group on Food Surveillance
The Working Party on Styrene

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INTRODUCTION

1. Styrene has been known since the eighteenth century and was first synthesised in 1866, but it remained an obscure substance of merely academic interest until about 1925 when the polymer was first produced commercially in Germany. The large-scale production of styrene commenced during the Second World War and was brought about by the need to produce synthetic rubber, most of which was based on the copolymer styrene-butadiene. Since the late 1940s styrene has been employed extensively in the production of plastics for a wide range of applications including use as food contact materials. In several types of these plastics styrene is employed in combination with other monomers, such as acrylonitrile, although it is used as the sole monomer in the production of various types of polystyrene.

2. Until 1974 there were insufficient toxicological data available on styrene to enable an assessment of the long-term safety of styrene-containing materials for food contact purposes to be made.[1-24] Since that date, however, numerous studies have been carried out[25-131] including six major programmes of long-term toxicity testing, one on polystyrene itself. The tests were conducted at the Huntingdon Research Centre[25] (polystyrene), at Bologna University by Professor C Maltoni on behalf of a European consortium of companies[26], at the laboratories of the Dow Chemical Company in the USA on behalf of the US Chemical Manufacturers Association (CMA)[27,28], at the laboratories of Litton Bionetics Inc in the USA on behalf of the National Cancer Institute (NCI)[29] and of the CMA[30], and at the International Agency for Research on Cancer (IARC) in France.[31] Several occupational studies on exposed employees have been carried out on styrene.[3,19,21,37-49]

3. In view of the considerable toxicological interest being shown in styrene and, in particular, because of a lack of information on the potential exposure to this substance from the ingestion of food, the Ministry of Agriculture, Fisheries and Food considered it necessary to obtain the following information:

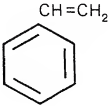
(a) the food contact materials in which styrene is used,
(b) the food contact uses of these materials,
(c) the level of monomer throughout the 'food packaging chain',
(d) the factors affecting the monomer level in the material and in food with which it has been in contact,
(e) the likely daily intake of styrene from food.

It was recognised at an early stage that realisation of the objectives of this investigation would require close collaboration with Industry. The Working Party on Styrene was, therefore, established with a membership drawn largely from producers and users of styrene polymer or copolymer food contact materials or articles and from food manufacturers. The first meeting of the Working Party took place in October 1979.

Manufacture and Use of Styrene

4. Styrene (vinylbenzene, ethenylbenzene) is a colourless, mobile liquid with a penetrating odour and which is miscible with the common organic solvents but only sparingly soluble in water. On exposure to light and air it slowly polymerises

and undergoes oxidation with the formation of peroxides. Styrene has the chemical structure shown below:



The commercial manufacture of styrene is normally carried out by the catalytic dehydrogenation of ethylbenzene, which may itself be produced with high purity by the direct alkylation of benzene with ethylene. Styrene is recovered from the reaction mixture by distillation under reduced pressure in the presence of suitable non-volatile polymerisation inhibitors.

5. In the United Kingdom styrene is used in the production of a number of polymers and copolymers which have a wide range of food contact applications. The manufacture of the copolymers acrylonitrile-butadiene-styrene (ABS) and styrene-acrylonitrile (SAN) has already been described in some detail in a previous report in this series.[132] Furthermore, several of the other copolymers of styrene are of only limited interest in this Survey, either because their usage in the UK is very small or because they are used predominantly in applications which involve only indirect contact with food. It is not appropriate in a report of this nature to give detailed consideration to the methods of manufacture of all of the many polymers and copolymers of styrene and, therefore, discussion will be restricted to those of major importance which have not already been considered elsewhere.

Manufacture of Styrene Polymers and Copolymers

6. The unmodified or general purpose grade of polystyrene (PS) is produced either by a continuous mass process or by a suspension process. In the manufacture of PS by the continuous mass process, styrene, optionally mixed with a non-polymerisable volatile diluent, is passed successively through two or more reactors each of which is equipped with several heat exchange zones and an agitator. The reaction mixture containing typically about 85% PS together with unreacted monomer and any diluent is then transferred to a low pressure, high temperature devolatilisation tower where the unreacted monomer and diluent are removed and recycled. The hot polymer is fed directly into an extruder from which the polymer strands are cooled and cut into pellets. In the suspension process, tiny droplets of styrene are dispersed in water in the presence of a small quantity of a suspending agent (typically 0.01 – 0.05% of the monomer mass) and a polymerisation initiator, which is usually a mixture of organic peroxides. The reaction mixture is heated in a reactor equipped with an agitator until polymerisation is substantially complete. The polymer beads are washed, centrifuged, dried and finally pelletised after passage through an extruder. Residual monomer levels in pellets produced by either process may be substantially reduced by means of appropriate vacuum-stripping techniques or by the use of a scavenger catalyst. The pellets are usually converted into end products either by injection moulding or by sheet extrusion followed by thermoforming.

7. Expanded polystyrene (EPS) is fabricated from polystyrene containing an expanding agent (a substance of low boiling point such as pentane) and a nucleating agent, using one of two processes. In the first process, commonly used in the production of trays, PS pellets are extruded into sheet, the expanding agent being injected under pressure into the molten polymer in the extruder. The expanded PS sheet is subsequently thermoformed. In the second process, which is used for most other applications, PS beads containing the expanding agent are treated with steam to expand them to the desired bulk density and then left to stand for about 16 hours. The intermediate 'prefoam' produced in this way is subsequently placed in a mould and once again treated with steam to produce the final article.

8. The high impact grade of polystyrene (HIPS) like the general purpose grade PS can be produced using either a continuous mass process or a suspension process but in this instance a small proportion (typically 3–10%) of a rubber (usually polybutadiene) is dissolved in the styrene monomer prior to polymerisation. In the suspension process, the styrene solution of the rubber is normally polymerised in mass to a conversion of 20–30% prior to formation of the suspension.

9. Another important use of styrene is in the production of glass reinforced plastic (GRP). Styrene is employed as a cross-linking agent with polyester resins to produce thermosetting, insoluble and infusible products; these cross-linked resins are brittle and, in consequence, are almost always reinforced with glass-fibre. The traditional fabrication process involves the synthesis of an unsaturated polyester resin which is mixed with styrene, the quantity added ranging from 30–50% of the resin depending on the degree of cross-linking required. The resin is mixed with a catalyst (for example 0.4% by weight of cobalt naphthenate) and spread on glass fibre reinforcement using a hand-held roller. It is then left to cure at room temperature for 24 hours and post-cured at 80° – 85°C for several hours to reduce levels of residual styrene.

Legislative Controls

10. There are no specific regulations controlling the presence of styrene in food in the United Kingdom. However, the general provisions of the Food and Drugs Act 1955 make it an offence in England and Wales to sell or expose for sale to the consumer, food which has been rendered injurious to health (Section 1); which is not of the nature, substance or quality demanded (Section 2) or which is unfit for human consumption (Section 8). Regulations could also be made under the Consumer Safety Act 1978 to control the quantity of styrene in plastics articles intended for use with food. Furthermore the Materials and Articles in Contact with Food Regulations 1978 (as amended) require that materials and articles to which the Regulations apply shall be manufactured in accordance with good manufacturing practice, i.e. in such a way that under normal or foreseeable conditions of use they do not transfer their constituents to foods with which they are, or are likely to be, in contact, in quantities which could endanger human health or bring about a deterioration in the organoleptic characteristics of such food or an unacceptable change in its nature, substance or quality (Regulation 4(2)). The situation in Scotland and Northern Ireland is slightly different in that

these countries have their own Food and Drugs Acts. The Materials and Articles in Contact with Food Regulations 1978 apply in Scotland and similar arrangements apply in Northern Ireland.

THE SURVEY

Food Contact Uses of Styrene Polymers and Copolymers

11. The food contact uses of the various styrene polymers and copolymers in the UK are diverse and include packaging materials, kitchenware and household appliances, adhesives, can sealants and cargo and storage containers. A list of the various types of styrene polymer and copolymer food contact materials, together with examples of the food contact usage of each of these is set out in Appendix I. The approximate quantities of these materials used in the UK during the most recent year for which data are available are given in Appendix II.

(a) *Polystyrene (PS)*

12. The unmodified or general purpose grade of PS, which is also known as crystal PS, is a glass-clear, hard polymer with good electrical properties, easy processing characteristics and a high resistance to acids, alkalis and the lower alcohols. However, it also possesses poor gas barrier properties and for many purposes the impact strength is not considered adequate. Nevertheless, it is relatively inexpensive and finds use in a wide range of applications where toughness is of secondary importance, such as plates, drinking goblets and dessert tubs. To overcome the problem of inherent brittleness associated with PS several modified forms have been developed. These include HIPS (see paragraph 14) and oriented PS. Oriented PS is the product formed when PS is biaxially oriented, a process which improves the impact strength and toughness of the polymer while maintaining gloss and transparency. Currently the limited use of this material is in the film for cake box windows and for the protection of light-weight glass bottles.

(b) *Expanded Polystyrene (EPS)*

13. EPS exhibits excellent resistance to bacterial and mould growth and is virtually impervious to water, grease and oil. It is also very light, non-abrasive and, because of its cellular structure, possesses excellent cushioning properties and low thermal conductivity. These various properties combined with its low cost and ease of production have resulted in its widespread use for packaging certain types of food products. These include fragile foods such as eggs and fresh fruit, hot foods from 'take-away' restaurants and foods transported in a frozen or cold state such as fresh fish. In addition there is some use of EPS cups in vending machines.

(c) *High Impact Polystyrene (HIPS)*

14. The incorporation of various rubbers in PS is extensively used for improving the impact strength of the material and such grades are used for numerous packaging applications. A number of rubbers have been examined but the one of

choice is normally polybutadiene and, depending on the proportion incorporated, materials with varying degrees of impact strength can be produced. The polybutadiene serves to prevent crack propagation and increases extensibility although with some loss of transparency and tensile strength. The polymer is usually converted into the final product by extrusion and thermoforming although it may also be processed using other techniques such as injection moulding. HIPS articles may be produced by the converter from HIPS granules alone or from a blend of HIPS granules and PS granules. The advantage of the latter process is that a converter need only purchase a limited number of grades of HIPS and, by blending these with PS in differing proportions in-house, he is able to produce articles with varying degrees of impact resistance whilst minimising expenditure on plant and storage facilities. The use of blends is particularly common where articles are produced by thermoforming, it is less common when injection moulding is employed. HIPS is a relatively inexpensive product and so has found wide application in the packaging of a large number of food commodities, for example containers for dairy products (yogurt, cream etc), tubs for a wide variety of foods and vending cups. One developing use where it is currently the preferred material is in the fabrication of tubs for the packaging of aseptic products which have an increased shelf life. A recent trend has been towards the use of HIPS/PS laminates to produce food tubs. In this application the food contact layer is invariably HIPS and this is coated with a thin exterior layer of PS to enhance the appearance. This type of laminate is also used in the production of domestic appliances and refrigerator linings. Food tubs fabricated from a laminate comprising HIPS food contact and exterior layers with a polyvinylidene chloride layer sandwiched between to enhance the barrier properties have also been developed, but these are only used to a very small extent.

(d) *Acrylonitrile-Butadiene-Styrene (ABS) and Styrene-Acrylonitrile (SAN)*

15. The terpolymer ABS combines the properties of toughness, rigidity and resistance to many chemicals and solvents with ease of processing by injection moulding, or by extrusion and subsequent thermoforming. ABS has a high resistance to environmental stress cracking and its major packaging use in the UK is in the fabrication of tubs for soft margarine. ABS tubs are also used for packaging other types of foods and the terpolymer is employed in other types of food packaging besides tubs, in refrigerator linings, in kitchen appliances such as food mixers and in the production of piping for use in the food industry. SAN exhibits the useful properties of clarity, resistance to chemical attack and rigidity. It is relatively simple and inexpensive to process by injection moulding and is used for internal trays and fittings in refrigerators, household utensils such as coffee percolators and for luncheon boxes.

(e) *Glass Reinforced Plastic (GRP)*

16. GRP is used extensively in the fabrication of large bulk containers for both the transportation and storage of a wide range of foodstuffs. The main advantage of this material is its strength combined with low corrosion characteristics and very light weight. In addition, vessels with a very large capacity (several thousand gallons) may be produced in one piece. GRP pipework is also widely used for the conveyance of both powdered and liquid foodstuffs.

(f) *Other uses*

17. The copolymer methyl methacrylate-butadiene-styrene (MBS) is added at a level of about 10% to PVC which is then used in the fabrication of bottles, films, food containers and cups. MBS significantly enhances the impact resistance properties of the PVC containers. Styrene-butadiene copolymer (containing 75% styrene) which is used for the fabrication of cups, tubs and trays is a relatively recent innovation. This copolymer, which is often used as a 50 : 50 blend with PS, may be employed as a direct replacement for HIPS. It possesses the advantage of crystal clarity but it is also more expensive at the present time. Styrene-butadiene rubber (containing 25 – 50% styrene) on the other hand, has been known for many years. This is widely used as a can sealant and also employed in the manufacture of grommets, seals and conveyor belting intended for food contact. Finally, a range of styrene copolymers are used as hot melt adhesives and as coatings for paper and board. However, such coatings are used to improve printability and are, therefore, normally applied only to the non-food contact surface.

(g) *Survey of the Food Contact Applications of Styrene Polymers and Copolymers*

18. As has already been mentioned, in the United Kingdom a wide range of foods is either packaged or otherwise comes into contact with the various polymers and copolymers of styrene. The Working Party has undertaken a comprehensive survey of the food contact applications of styrene polymers and copolymers and the findings are set out in Appendix III. It can be seen from these data that the most important usages of styrene polymers and copolymers, from the point of view of food contact applications, are in the fabrication of tubs for a wide range of foods, of trays for fresh meat, fresh fruit and frozen fish and of cups.

Analysis for Styrene

19. Several methods of analysis for styrene in food contact materials and in foods were identified and/or developed, and extensively tested in collaborative studies by the Working Party. In the case of most styrene polymers and copolymers it was found that the residual styrene could be determined in a satisfactory manner by gas chromatography (gc) using a flame ionisation detector (FID). The injection of either an aliquot of a solution of the polymer or copolymer in a suitable organic solvent, for example dimethylformamide (DMF) or dimethylacetamide, or of the headspace from above a heated equilibrated solution of the polymer or copolymer[133], could be employed. The direct injection technique possessed the advantage of speed, as it did not require an equilibration step, but the headspace technique was capable of achieving higher sensitivity and better reproducibility. For most polymers and copolymers the limit of detection using the headspace techique was found to be 1 mg/kg and using the direct injection technique 50 to 100 mg/kg, although 10 mg/kg could be achieved by an experienced analyst. The determination of the styrene content of GRP was less convenient owing to the heterogeneous nature of this material. It was therefore found necessary to devise a statistical sampling procedure to ensure that a representative portion of GRP was taken. This portion was then heated in a sealed vial with DMF until the plastic had fully dissolved and, after equilibrium had been reached, the headspace was sampled automatically and injected into the gas chromatograph. The limit of detection for this procedure was approximately 10 – 20 mg/kg.[134]

20. For the quantification of styrene in food it was found that headspace gc using an FID did not have a sufficiently low limit of detection. Thus it was necessary either to preconcentrate the styrene by distillation of an aqueous or methanolic slurry of the food, extracting the distillate with hexane and injecting the hexane extract into a gas chromatograph equipped with an FID[135, 136] or to employ a headspace technique using a mass spectrometer in the selected ion mode as a more sensitive detector than the FID.[137] Using either of these techniques it was possible to achieve a limit of detection of 1 µg/kg for the majority of foods and 10 µg/kg for margarine and other table spreads. All samples of food were homogenized prior to analysis.

RESULTS

Styrene Content of Food Contact Materials

21. Many data have been collected by the Working Party on the levels of residual styrene in the various styrene polymer and copolymer food contact materials, both before and after contact with food. This information is summarised in Appendix IV.

22. The information set out in Table 1 of Appendix IV comprises, for some of the most important styrene polymer and copolymer food contact materials, historical results obtained from a number of converters. It is apparent, that although there is a considerable variation in levels over the years, there has been an overall reduction during the last ten years. However, most manufacturers still offer a range of polymers and copolymers each with a different residual styrene level. The most pronounced reduction in residual styrene levels has been achieved in the fabrication of GRP. In 1969 levels of 3–7% residual styrene in GRP were occasionally encountered and such levels were also found to be associated with instances of taint. This problem was quickly recognised and a major effort made to reduce substantially levels of residual styrene. Today using modern post-curing techniques, a level in the order of 20–200 mg/kg residual styrene may be achieved. Furthermore, the British Plastics Federation has instituted a Code of Practice which limits the level of total volatiles in GRP to a maximum of 1,000 mg/kg. However, problems with taint are still encountered occasionally in foods transported in GRP containers that have been manufactured in other countries. In Table 2 of Appendix IV current typical and maximum levels of residual styrene in styrene polymer and copolymer food contact materials and articles are detailed. In most polymers or copolymers a level of about 200 mg/kg residual styrene is believed to be an irreducible minimum as the polymer or copolymer is susceptible to thermal degradation to a certain extent during processing.

23. During 1980 a major investigation was undertaken by one converter into the effect of thermoforming on the residual styrene content of HIPS and PS granules. The results of studies involving the processing of a 50:50 blend of PS/HIPS granules into either cups or containers for dairy products are set out in Tables 3 and 4 of Appendix IV, respectively. It is apparent from these studies that levels of residual styrene tend to fall slightly during thermoforming. Details have been obtained from a number of converters regarding the levels of residual styrene in styrene polymer and copolymer granules, and in the articles manufactured therefrom, and these are set out in Table 5 of Appendix IV. These results afford

further evidence that there is a general tendency for residual styrene levels to fall during thermoforming. However, in contrast, levels appeared to rise slightly during injection moulding. It was thought that this latter effect was probably caused by slight thermal decomposition owing to the higher polymer temperature normally employed during injection moulding.

24. During the early part of 1981 the Working Party undertook a wide-ranging survey of styrene levels in retail foods and their containers. The results of this study, which encompassed the majority of those foods that were known to be packed in containers fabricated from styrene polymers or copolymers, are set out in Table 6 of Appendix IV. It can be seen from this Table that, although levels of up to 1400 mg/kg residual styrene were occasionally found, for all of the food containers examined the mean level of residual styrene in every case was less than 720 mg/kg.

Styrene Content of Foods

25. Those foods which might be expected to make a significant contribution to the total dietary intake of styrene were identified by the Working Party. All of these foods were included in the analytical survey of retail samples, the results of which are set out in Table 6 of Appendix IV. The majority of these foods were types which are normally eaten after purchase without further preparation and these were simply removed from their containers and homogenized prior to analysis. The 'take-away' foods (hamburger and sweet and sour sauce) and the beverages (coffee, tea, lemon tea, chocolate, soup and fruit drinks from a number of different vending machines) were left in their containers for 30 minutes, which was considered to be the longest realistic storage period, prior to removal for analysis. The samples of fresh meat and frozen fish were cooked by simmering in water for 30 minutes before analysis, as this was considered to be the gentlest method of cooking and therefore the least likely to drive off any styrene which might be present. Both the homogenized food and the cooking liquor were analysed.

26. Most of the foods examined which had come into contact with styrene polymers or copolymers were found to contain a measurable quantity of styrene. The levels of styrene recorded in individual samples ranged from < 1 µg/kg to 205 µg/kg and both of these extremes were found in chilled desserts. The mean levels recorded ranged from 2 µg/kg in preserves to 26 µg/kg in yogurt. The higher levels of styrene in food occurred only rarely and were often associated with high levels of styrene in the container. However, this was not always the case and the level of styrene in a food could not be related to any one particular factor. Levels varied for instance with the nature of the food, the level of residual styrene in the container and the length and temperature of storage. Laboratory studies carried out in 1970 to investigate the factors affecting the migration of styrene from food contact materials into food indicate that factors influencing migration can be rubber content (of HIPS), storage temperature, and residual styrene content. In this study it was also observed that migration was lower into aqueous foods than into fatty foods.[138]

27. There is a possibility that styrene may be present in chewing gum as a result of the use of butadiene-styrene rubbers in chewing gum bases. Two forms of the

polymer, one comprising 75% butadiene/25% styrene and the other 50% butadiene/50% styrene, are listed in The Food Chemicals Codex, Third Edition, as being suitable for use in food. The specifications allow for up to 20 mg/kg and 30 mg/kg residual styrene, respectively, to be present in the polymers. It is believed that butadiene-styrene rubbers are not manufactured in the UK for use in chewing gum bases, though the polymers are present in some imported gum base used in the manufacture of finished gum. The Food Additives and Contaminants Committee is currently reviewing the components of chewing gum bases, and as the butadiene-styrene polymers are being evaluated in this Review, they will not be considered further in this Report.

Estimated Likely Intake of Styrene through the Diet

28. The likely intake of styrene per capita from the diet is very difficult to estimate for a number of reasons. The usage of styrene polymers and copolymers is so widespread that, at one time or another, the majority of foods are likely to come into contact with one or more of these. In addition, the conditions of contact may vary from only the briefest association to extended storage and, as was demonstrated by the retail survey conducted by the Working Party (Table 6 of Appendix IV), for any one food the content of styrene may vary over a wide range (for example from < 1 to 205 µg/kg for chilled desserts). However, although a large number of foods were identified as coming into contact with styrene polymers and copolymers, the Working Party was satisfied that, because of the low residual styrene contents of some packaging materials (for example MBS modified PVC), the brief periods of contact (for example HIPS lids used on containers fabricated from other materials) or the low consumption of certain foods (for example chocolate spread, glacé fruit), many would not make a significant contribution to the total per capita daily intake of styrene from the diet.

29. Those foods identified as being important in terms of their contribution to the dietary intake of styrene are listed in Table 6 of Appendix III together with the total tonnages packed per annum and the contribution of each to the average UK diet. Using this information together with that contained in Table 6 of Appendix IV on the mean and maximum levels of styrene in each food, it can be calculated that the average and maximum likely intakes per capita of styrene derived from the average UK diet are approximately 1 µg/day and 4 µg/day, respectively.

Health Considerations

30. In October 1982 an opinion was sought from the Department of Health Committee on Toxicity of Chemicals in Food, Consumer Products and the Environment (COT) on the possible health risks to man that might be created by the ingestion of small amounts of styrene from food contact materials. The Committee examined the toxicological and epidemiological evidence available at that time[1-131] and received advice from its sister committees, the Committees on Carcinogenicity and on Mutagenicity of Chemicals in Food, Consumer Products and the Environment, on the carcinogenicity and mutagenicity aspects, respectively. The COT concluded that, in view of the very low exposure to styrene from food as detailed in this Report, "there is no likely toxicological hazard to man from present levels of styrene in food". (See Appendix VI.)

CONCLUSIONS

31. The conclusions of the Working Party are as follows:

- (a) The food contact uses of styrene polymer and copolymer materials in the UK have been identified and are detailed in paragraphs 11 to 19 and Appendices I to III.
- (b) The levels of styrene in food contact materials and articles and in food have been determined and are detailed in Appendix IV.
- (c) The factors affecting the styrene content of food contact materials and articles and the migration of residual styrene into food have been investigated and are discussed in paragraphs 21 to 26.
- (d) The average and maximum likely per capita intakes of styrene from the average diet have been estimated and found to be approximately 1 μg/day and 4 μg/day, respectively (paragraphs 28 and 29).

32. The success of this investigation again demonstrates that joint action between Government and a responsible Industry is an effective means of collecting information to permit an assessment to be made of the significance to health of a contaminant in food.

33. The Committees on Carcinogenicity, on Mutagenicity and on Toxicity of Chemicals in Food, Consumer Products and the Environment have considered this Report together with the available toxicological and epidemiological information on styrene. The opinions of these Committees are set out at Appendix VI. The Food Additives and Contaminants Committee has also considered this Report and expressed the views which are recorded at Appendix VI.

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FOOD CONTACT USES OF STYRENE POLYMERS AND COPOLYMERS IN THE UK IN 1980

Food contact material	Comonomers used with styrene	Percentage by weight of styrene used in the manufacture of the polymer or copolymer	Examples of food usage	Other uses
Polystyrene (PS)	—	100%	Cups, tubs, trays, film, bottles, bottle caps, plates, crisper trays	—
Expanded polystyrene (EPS)	—	100%	Cups, trays, film, egg boxes, boxes for fish	—
High impact polystyrene (HIPS)*	Butadiene	85–98%	Cups, tubs, lids, trays, caps for board tubes, cutlery, sandwich boxes, closures for coffee jars, refrigerator linings	—
Acrylonitrile-butadiene-styrene (ABS)	Acrylonitrile, butadiene	50–75%	Tubs, lids, vacuum formed inserts, refrigerator linings, household appliances, ice lolly sticks, additive to PVC bottles and tubs, piping for the food industry	Impact modifier in PVC bottles
Styrene-acrylonitrile (SAN)	Acrylonitrile	70–80%	Cups, sandwich boxes, tumblers, picnic ware, cutlery, liquidisers, coffee percolators, crisper trays	—
Methyl methacrylate-butadiene-styrene (MBS)	Methyl methacrylate, butadiene	26–38%	Additive to PVC bottles, films, sheets, containers and cups	Impact modifier in PVC bottles
Styrene-butadiene block copolymer**	Butadiene	75%	Cups, tubs, trays	—
Glass reinforced plastic (GRP)	Methyl methacrylate, vinyl toluene (unsaturated polyester resin)	30–50% of resin	Pipes, silos, cargo containers, refrigeration units, containers for water storage in creameries, canneries and edible oil factories, for soaking peas and dried fruit, for sugar storage	Resin used as a binder with glass fibre

Styrene-acrylic copolymers	Butyl acrylate, acrylic acid	40–70%	Adhesive coatings for paper and binders for printing and coating on polyethylene and board	—
Styrene-butadiene-styrene Styrene-isoprene-styrene α-Methylstyrene-vinyltoluene resin	Butadiene Isoprene Vinyltoluene	20–30%	Hot melt adhesives for labels, tapes, case and carton sealing	—
Styrene-butadiene rubber	Butadiene	20–50%	Sealants in metal cans, grommets, seals and conveyor belting	—
HIPS/PS laminate	—	—	Refrigerator linings, household appliances, caps, tubs, lids	—
HIPS/polyvinylidene chloride/HIPS laminate	—	—	Tubs	—

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*HIPS articles may be produced either from HIPS granules alone or from a converter blend of HIPS granules and PS granules.

**Often used in a blend with PS.

APPENDIX II

QUANTITIES OF STYRENE POLYMERS AND COPOLYMERS USED FOR FOOD CONTACT IN 1980

Polymer or copolymer	Tonnage*
Polystyrene (PS)	—
Expanded polystyrene (EPS)	—
High impact polystyrene (HIPS)	—
Acrylonitrile-butadiene-styrene (ABS)	—
Styrene-acrylonitrile (SAN)	—
Methyl methacrylate-butadiene-styrene (MBS)	—
Glass reinforced plastic (GRP)	—
Styrene-butadiene rubber (SBR)	1,000
Other	<200

*Information on tonnages was made available to the Working Party by the British Plastics Federation on condition that this was treated in confidence.

SURVEY OF FOOD CONTACT APPLICATIONS OF STYRENE POLYMERS AND COPOLYMERS IN THE UK IN 1980

Table 1: Examples of food contact applications of polystyrene (PS)

Food commodity	Material or article	Type of food contact	Usual food contact conditions
Preserves, chocolate spread	Tubs	Direct	Ambient, up to 1 year
Ice cream	Tubs	Direct	−30°C, 1 to 10 weeks
Desserts	Tubs	Direct	11°C, up to 13 days
Glacé fruit	Tubs	Direct	Ambient, up to 1 year
Cakes, fruit	Trays	Indirect	—
Beer, spirits, soft drinks	Cups	Direct	Ambient, short periods
Vinegar	Bottles	Direct	Ambient, up to 1 year
Cakes	Film (for cake box windows)	Indirect	—
Dried herbs and spices	Caps (used with polypropylene tubs)	Direct but intermittent	Ambient, up to 1 year
Edible oils	Cap inserts	Direct but intermittent	Ambient, up to 1 year
Various	Plates	Direct	Various, short periods

Table 2: Examples of food contact applications of expanded polystyrene (EPS)

Food commodity	Material or article	Type of food contact	Usual food contact conditions
Eggs	Boxes	Direct	Ambient, 1−2 weeks
Confectionery	Film	Direct	Ambient
Fresh meat	Trays	Direct	5−10°C, 1 week
Takeaway foods	Trays and containers	Direct	Various, short periods
Various beverages	Cups	Direct	Ambient to 90°C, up to ½ hour
Fresh fruit and vegetables	Trays	Direct	Ambient, 1 week
Frozen fish	Trays	Direct	−12°C to −18°C, 1 week

Table 3: Examples of food contact applications of high impact polystyrene (HIPS)

Food commodity	Material or article	Type of food contact	Usual food contact conditions
Yogurt, cream, cottage cheese, desserts	Tubs	Direct	5 – 10°C, 1 – 2 weeks
Ice cream	Tubs and lids	Direct	– 30°C, 1 – 10 weeks
Coleslaw	Tubs	Direct	11°C, up to 17 days
Herbs and spices, dried onions, gravy mix, gravy salt	Tubs and lids	Direct	Ambient, up to 1 year
Imitation cream	Tubs	Direct	5 – 10°C, up to 3 weeks
Sugar spread, honey spread, chocolate spread	Tubs and lids	Direct	Ambient, up to 1 year
Glacé fruit, candied peel	Tubs	Direct	Cool, up to 1 year
Sodium bicarbonate, saccharin tablets	Lids (for carton-based drums)	Direct but intermittent	Ambient up to 1 year
Various beverages	Cups	Direct	Ambient to 90°C, up to 1½ hour
Cakes and biscuits	Trays	Direct	Ambient
Confectionery	Caps (for board tubes)	Direct but intermittent	Ambient, up to 1 year
Various	Cups, cutlery, sandwich boxes	Direct	Various, short periods
Coffee, non-dairy creamer	Closures for jars	Indirect	—

Table 4: Examples of food contact applications of acrylonitrile-butadiene-styrene (ABS)

Food commodity	Material or article	Type of food contact	Usual food contact conditions
Soft margarine, concentrated butter for cooking	Tubs and lids	Direct	Ambient/chill, 8 – 12 weeks
Nut products	Tubs	Direct	Ambient, up to 6 months
Sugar spread	Lids	Direct but intermittent	Ambient, 1 year or more
Biscuits and cakes	Vacuum formed inserts	Direct	Ambient
Various	Refrigerator linings, household appliances	Direct or indirect	Various
Various	Piping for the food industry	Direct	Various
Ice lollies	Sticks	Direct	– 30°C, 1 – 10 weeks
Various	Additive to PVC bottles and tubs	Direct	Various

Table 5: Examples of food contact applications of other styrene copolymers

Copolymer	Food commodity	Material or article	Type of food contact	Usual food contact conditions
Styrene-acrylonitrile (SAN)	Various	Utensils, household appliances	Direct or indirect	Various
Glass reinforced plastic (GRP)	Water, fruit juices, flour, baking ingredients	Pipes, silos, storage containers	Direct	Ambient
	Perishable foods, e.g. butter	Cargo containers, refrigeration units	Indirect	Low temperature, 8 – 10 weeks
Styrene-acrylic copolymers	Frozen chickens, bacon	Coated polyethylene film	Direct	Low temperature, extended storage
	Chocolate and other confectionery	Coated paper	Indirect	Ambient, several months
	Ice cream	Coated paper	Indirect	– 20°C, several months
	Nut products	Lids for ABS tubs	Direct but intermittent	Ambient, 6 months
Methyl methacrylate-butadiene-styrene (MBS) (impact modifier for PVC)	Aqueous foods (mineral waters, fruit juices, vinegar, wine etc)	Bottles, cups, containers	Direct	Ambient, up to 1 year
	Fatty foods (salad oils, margarine etc)	Bottles, cups, containers	Direct	Ambient, up to 1 year
	Fresh meat, fish	Film, sheet, containers	Direct	Low temperature
Styrene-butadiene rubber	Canned foods and beverages	Can sealant	Indirect	—
Styrene-butadiene-styrene Styrene-isoprene-styrene α-Methylstyrene vinyl toluene resin	Various	Hot melt adhesives for labels, tapes, case and carton sealing	Indirect	—
50 : 50 Blend of styrene and styrene-butadiene copolymer	Desserts, cake decorations, coleslaw	Tubs	Direct	Various
	Cold beverages	Cups	Direct	Low temperature
	Soft fruit, vegetables	Trays	Direct	—
PS/HIPS laminate	Various	Refrigerator linings, household appliances	Direct or indirect	Various
	Various	Cups, tubs, lids	Direct	Various
HIPS/polyvinylidene chloride/ HIPS laminate	Various	Tubs	Direct	Various

Table 6: Quantities of the major food commodities packed in direct contact with styrene polymers or copolymers

Food commodity	Tonnage/annum	Contribution to the average UK diet (g/person/day)
Beverages (including those from vending machines)	1,090,000	53.3
Fresh meat	430,000*	21.0
Margarine	158,000	7.7
Yogurt	113,000	5.5
Frozen fish	45,000*	2.2
Fresh fruit	45,000*	2.2
Cream	22,000	1.1
Ice cream, chilled desserts	22,000	1.1
Coleslaw, prepared salads	17,000	0.8
Soft cheese	16,000	0.8

*Estimated

LEVELS OF RESIDUAL STYRENE IN FOOD CONTACT MATERIALS

Table 1: Historical levels of residual styrene in styrene polymer and copolymer pellets

	Residual styrene (mg/kg)*									
	Polystyrene (PS)		Expanded polystyrene‡ (EPS)		High impact polystyrene (HIPS)		Acrylonitrile-butadiene-styrene (ABS)		Styrene-acrylonitrile (SAN)	
Year	Mean	Maximum	Mean	Maximum	Mean	Maximum	Mean	Maximum	Mean	Maximum
1968	—	—	—	—	920; 1410	1400; 1800	—	—	—	—
1969	—	—	—	—	860	1930	—	—	—	—
1970	—	—	—	—	1520; 1720	3250; 3680	—	—	—	—
1971	—	—	—	—	1520; 800	2000; 1300	—	—	—	—
1972	—	—	—	—						
1973	1030; 2400	—	—	—	920	1460	—	—	—	—
1974	—	—	—	—	—	—	1300**	2400**	—	—
1975	1670	3500	1500	—	—	—	900**	2200**	—	—
1976	870	970	—	—	800	1270	700**	1600**	—	—
1977	700; 800	1020; 1100	—	—	600	990	300**	1060**	3400	5000
1978	380	580	1400	—	420	840	300**	800**	1000	1550
1979	400	790	1400	—	380	600	300**; 600†; 890††	700**; 790†; 1220††	950	1300
1980	410	600	1000	—	360	490	300**; 600†; 700††	600**; 1000†; 870††	950	1250

*Results obtained from a number of different converters. Each result refers to a specific converter.

**Intended for food tubs. †Intended for refrigerator applications. ††Intended for household appliances. ‡Level in expandable polystyrene beads.

Table 2: Levels of residual styrene in styrene polymer and copolymer food contact materials and articles in 1980

Polymer or copolymer	Residual styrene (mg/kg)	
	Typical	Maximum
Polystyrene (PS)	300 – 1000	2500
Expanded polystyrene (EPS)	300 – 1000	2000
High impact polystyrene (HIPS)	300 – 1000	2500
Acrylonitrile-butadiene-styrene (ABS) – for food tubs	200 – 300	600
Acrylonitrile-butadiene-styrene (ABS) – other uses	300 – 1000	2000
Styrene-acrylonitrile (SAN)	600 – 1200	2000
Methyl methacrylate-butadiene-styrene (MBS)	not detectable – 10	30
Styrene-butadiene block copolymer	—	—
Glass reinforced plastic (GRP)	20 – 200	1000
Styrene acrylic copolymers	60 in latex	—
Styrene-butadiene-styrene Styrene-isoprene-styrene α-Methylstyrene-vinyltoluene resin	Not used for direct food contact purposes	
Styrene-butadiene rubber – raw polymer	10 – 30	
– cured polymer	< 1	1

Table 3: Effect of processing on residual styrene levels in HIPS/PS granules – production of cups*

	Residual styrene (mg/kg)		
	PS granules	HIPS granules	Cups
Mean	430	350	340
Standard deviation	72	68	57
Maximum	580	510	500
Minimum	340	230	230
Number of samples	10	43	33

Table 4: Effect of processing on residual styrene levels in HIPS/PS granules – production of food containers*

	Residual styrene (mg/kg)		
	PS granules	HIPS granules	Food containers
Mean	390	390	360
Standard deviation	91	73	59
Maximum	570	570	480
Minimum	250	220	230
Number of samples	61	60	60

*Studies carried out during May-August 1980 using a 50 : 50 mixture of PS/HIPS granules. Cups and food containers produced from the granules by extrusion and subsequent thermoforming.

Table 5: Effect of processing on residual styrene levels in styrene polymers and copolymers

Polymer or copolymer	Nature of final article	Method of processing	Number of samples	Residual styrene (mg/kg)			
				Granules		Final article	
				Range	Mean	Range	Mean
HIPS	Food containers	Injection moulding	12	600 – 800	708	700 – 1000	833
HIPS	Food containers	Thermoforming	3	390 – 700	493	320 – 700	450
HIPS	Cups	Thermoforming	3	430 – 800	593	350 – 800	567
HIPS	Cutlery	Injection moulding	1	—	470	—	580
HIPS	Refrigerator lining	Thermoforming	1	—	630	—	700
PS	Cups	Injection moulding	3	490 – 610	557	550 – 700	640
PS	Food container	Injection moulding	1	—	1300	—	1070
PS	Crisper tray	Injection moulding	1	—	770	—	810
PS	Cutlery	Injection moulding	1	—	750	—	800
EPS	Cups	Steam moulding	2	930 – 1020	975	330 – 650	490
EPS	Trays	Thermoforming	2	340 – 510	425	360 – 500	430
ABS	Food containers	Thermoforming	24	150 – 500	315	120 – 400	246
ABS	Flour shaker	Injection moulding	1	—	380	—	370
SAN	Coffee percolator	Injection moulding	1	—	820	—	850

Table 6: Retail survey of foods and their containers*

	Container			Food		
	No. of container samples analysed	Residual styrene (mg/kg)		No. of food samples analysed	Residual styrene (μg/kg)	
Food commodity		Container – range	Container – mean		Food – range	Food – mean
Coleslaw, prepared salads	25	60 – 550	351	24	< 1 – 20	7
Cream	21	280 – 1390	646	20	< 1 – 70	11
Soft cheese	24	140 – 1220	494	23	< 1 – 125	16
Dessert products	17	50 – 650	303	16	< 1 – 205	22
Yogurt	40	280 – 1400	565	34	< 1 – 200	26
Margarine, table spreads	27	90 – 390	243	27	< 10 – 10	10
Chocolate spread, honey spread, preserves	7	300 – 530	399	7	< 2 – 2	2
Beverages	20	340 – 1040	633	18	< 2 – 45	12
Fresh meat	4	330 – 430	377	4	Meat: 1 – 5** Liquid: < 1	Meat: 3 Liquid: < 1
Fish	4	230 – 470	333	4	Fish: < 1 – 1** Liquid: < 1	Fish: 1 Liquid: < 1
Fresh strawberries	5	430 – 1120	718	5	< 3 – 10	5
Eggs	3	430 – 510	460	3	< 1	< 1
Glacé fruit	2	120 – 920	520	1	5	5
Takeaway food	3	20 – 340	197	3	< 1 – 5	2
Wine	—	—	—	3	< 1	< 1

*The analyses were undertaken on behalf of the Working Party by a number of industrial laboratories and the Ministry's Food Laboratory (Norwich).

**Analyses carried out on cooked foods.

APPENDIX V

ORGANISATIONS CONSULTED DURING THE COURSE OF THE SURVEY

British Adhesive Manufacturers' Association
British Association of Synthetic Rubber Manufacturers
British Carton Association
British Fibreboard Packaging Association
British Paper and Board Industry Federation
British Plastics Federation
British Resin Manufacturers' Association
British Rubber Manufacturers' Association Ltd
Chemical Industries Association Ltd
Creamery Proprietors' Association
Expanded Polystyrene Association
Flexible Packaging Association
Food and Drink Industries Council
Food Manufacturers' Federation Inc and affiliates
Metal Packaging Manufacturers' Association
Paintmakers' Association of Great Britain Ltd
Process Plant Association
Shipowners' Refrigerated Cargo Research Association
UK Provision Trade Federation

CONSIDERATION OF STYRENE BY EXPERT COMMITTEES

COMMITTEE ON TOXICITY OF CHEMICALS IN FOOD, CONSUMER PRODUCTS AND THE ENVIRONMENT

Report on Styrene

Introduction

The COT last considered the safety-in-use of styrene as a monomer in the production of food contact plastics in 1978. At that time the COT agreed that no final decision on styrene was possible on the basis of the data then available. In addition to the results of long-term feeding studies on styrene which were then underway, the COT also expressed the wish to see information on food uses of polystyrene, residual levels of styrene and the amount of styrene present in contained food.

The Working Party on Styrene was established to obtain such information; its Report includes estimates of the average and maximum likely per capita intakes of styrene from the average diet, as a result of its wide range of food contact applications, of approximately 1 and 4 μg/day respectively.

Toxicology

Styrene administered orally to rats is excreted rapidly in the urine.[93] Tissue distribution studies indicate that the kidney has the highest concentration, although fat is also important.[53, 93, 113] Styrene oxide would appear to be a short-lived metabolite. In man the metabolic pathways seem to be similar to that of animals, the main urinary metabolites being phenyl glyoxylic and mandelic acids.[54] Styrene and its metabolites are found in the blood and urine of workers exposed to approximately 10 ppm styrene[48] but it is unlikely to be accumulated in the body of workers, even when inhaled repeatedly for 8 hours per day.[95, 119]

Haemosiderin was detected in liver reticuloendothelial cells of dogs dosed orally with very high levels of styrene (400 and 600 mg/kg/day) for 19 months.[28] However no such effect was seen in a 2-year rat feeding study.[30]

Reproduction studies revealed a trend towards reduced numbers of fetuses per litter but no teratological effects were seen.[73] In the rabbit delayed ossification was seen following exposure to 600 ppm styrene for 7 hours/day on days 6 – 18 of gestation.[85] The three-generation reproduction study in the rat revealed no treatment-related effects.[30]

There were some EEG changes in workers exposed to 30 ppm styrene for 8 hours.[125] Visuomotor accuracy was reduced following exposure to 55 ppm styrene for 8 hours. Tiredness and impaired reaction times have been found at worker exposure levels of 100 ppm styrene.[39] Slight hepatic effects, as judged by raised plasma enzyme levels, were also noted.[43]

Data on the mutagenicity and carcinogenicity of styrene were referred to the Committees on Mutagenicity and Carcinogenicity respectively (see below).

The COT is also aware of the recent report from the EC Scientific Committee for Food (Reports of the Scientific Committee for Food, 13th Series 1982).

Conclusions

The exposure of man to styrene in food from the migration from food contact materials is very low. Biological data indicate that man eliminates styrene effectively even at levels significantly in excess of human exposure from food. Styrene itself is not mutagenic and although styrene oxide is mutagenic when tested directly, its transient production during the metabolism of styrene *in vivo* is not thought to be significant for man in the context of exposure to styrene from food.

There is no convincing evidence that styrene is carcinogenic in animals and the currently available epidemiological data do not indicate that styrene is a human carcinogen.

Although relatively high exposure levels of styrene in man may lead to hepatic and neurotoxic effects, there is no likely toxicological hazard to man from present levels of styrene in food. However, the recent trend of reducing styrene monomer levels in polymers containing styrene should be encouraged.

COMMITTEE ON MUTAGENICITY OF CHEMICALS IN FOOD, CONSUMER PRODUCTS AND THE ENVIRONMENT

The Committee reached the following conclusions on the Report of the Working Party on Styrene:

1. Styrene itself is not a mutagen.
2. Evidence from tests for mutagenic effects in workers exposed occupationally to styrene is reassuring.
3. Styrene oxide when tested directly is mutagenic – but the transient production of styrene oxide, during the metabolism of styrene *in vivo* is not thought to be significant in man.
4. The very low levels of styrene found in food from the use of food contact materials do not represent a mutagenic hazard for man.

COMMITTEE ON CARCINOGENICITY OF CHEMICALS IN FOOD, CONSUMER PRODUCTS AND THE ENVIRONMENT

The conclusions reached by the Committee on the Report of the Working Party on Styrene were:

1. There is no convincing evidence that styrene is carcinogenic in animals.
2. Epidemiological evidence now available does not indicate that styrene is a human carcinogen.
3. The available evidence suggests that the very low levels of styrene found in food, from the migration from food-contact materials, do not indicate a hazard for humans.

FOOD ADDITIVES AND CONTAMINANTS COMMITTEE

The Food Additives and Contaminants Committee has considered this Report and endorses the comments of the COC, COM and COT. Furthermore the Committee welcomes the reduction in residual styrene levels already achieved and expects efforts to be continued in this direction.

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